#### black carbon

# from combustion source to radiative impact ...where are the biggest uncertainties?

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14 October 2004
Black Carbon Emissions & Climate Change
San Diego, California

# acknowledgements - thanks

- Workshop organizers! for invitation & focus
- Collaborators

Haolin Sun, UIUC; Bob Bergstrom, NASA-Ames; David Covert, U Washington; David Streets, Argonne Nat'l Labs; Trish Quinn & Tim Bates, NOAA-PMEL; Phil Rasch, NCAR

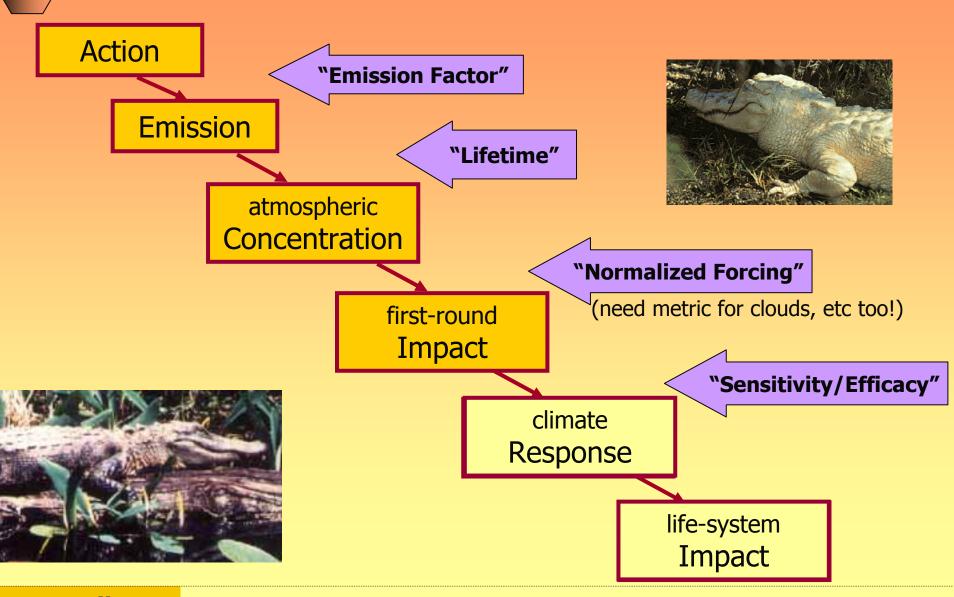
- Funding & Support
  - U.S. EPA: STAR grant; Climate office
  - NASA: ACMAP & EOS
- → Offsets for travel CO<sub>2</sub>
  - travel.500ppm.com



# outline

- 1. a Trail, 2 Hypotheses, and a (re)Definition
- 2. Inventories
- 3. Global models, a very little
- 4. Climate-relevant properties
- 5. finale: Tami's Top 4

## the Trail (beset by Alligators)



1. a Trail, etc.



#### the purpose of climate modeling

is to go where measurements cannot:

the past, the hypothetical future, the big (global) and little (microphysical) pictures

- OK for assessment of present:
  - generalized emissions (no explicit action linkage)
  - observationally-based forcing only
- need more direct linkage for...
  - past/future modeling



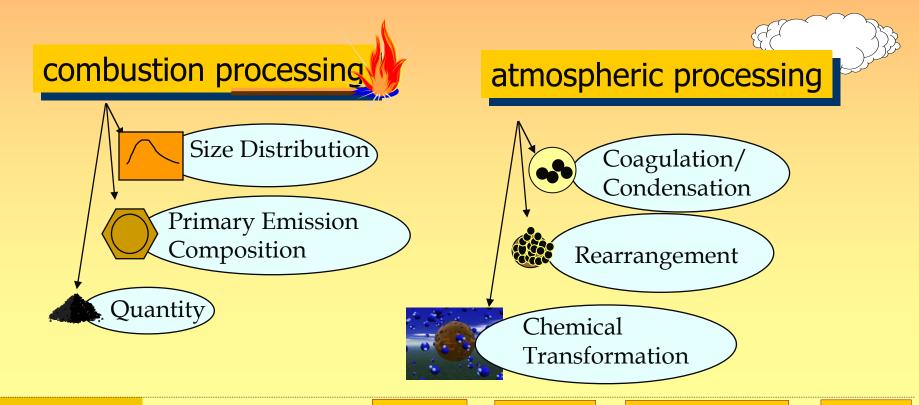
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1800



two distinctly different environments

influence the properties that govern aerosol-climate interaction



1. a Trail, etc.

action → emission -

concentration

impact



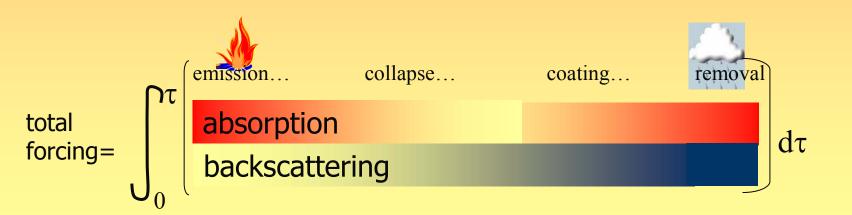
### definition: emission inventory

#### the expanded concept:

a tabulation of particle quantities and properties governed by the "combustion domain"

source-dependent, climate-relevant characteristics

our goal: estimate total impact of technology change



1. a Trail, etc.

action → emission

concentration

impact

# global inventories: "Bond/Streets"

BC emission = fuel use x PM emission factor x characteristics

- activity levels (usually *fuel*)
  - International Energy Agency, United Nations, etc.
- technology divisions
  - division into >100 fuel+technology categories
  - regionally-distinct technology divisions
- emission factors (PM x BC fraction)
- gridding
  - by population, land use, fire counts
- → management: SPEW (Speciated Pollutant Emission Wizard)
  - relational database (~40 descriptive linked databases)

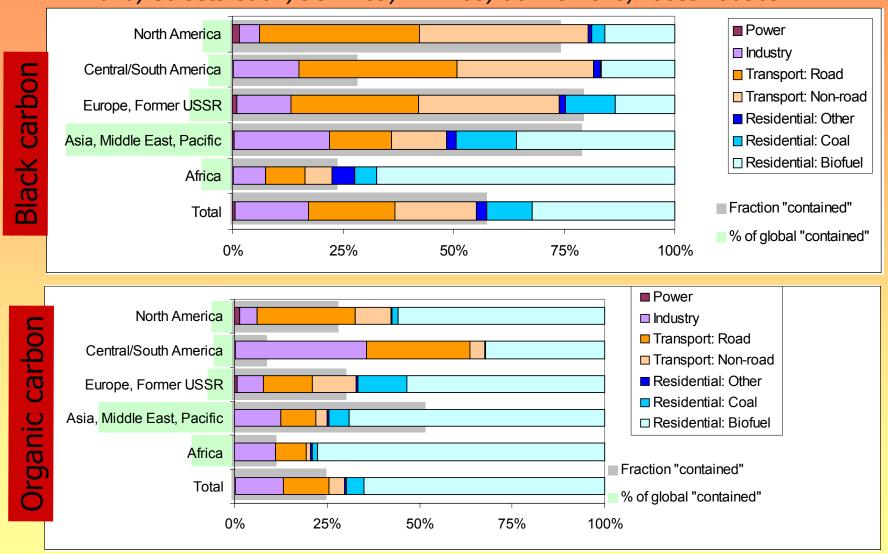
perturbations: China-BC (2001), TRACE-P (2003), future (2004) & past (in progress) estimates

## global inventories: state of the science

- → spatial resolution: 1°x 1°
  - sub-grid processes must be represented separately (and usually aren't)
- aerosol composition: minimal
  - last IPCC: OC=4xBC for fossil fuel
  - new: activity-specific OC+BC
- aerosol physical state: rudimentary
  - most: old/unsupportable values for size/optics

# global sources

Bond, Streets et al., JGR 109, D14203, doi:10.1029/2003JD003697



action

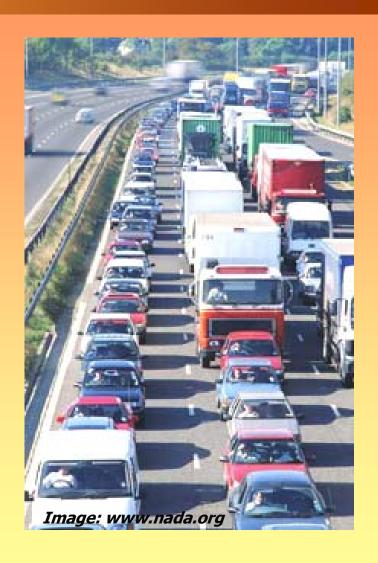
emission

concentration

impact

### transport sector treatment

- + on-road
  - estimated fleet emission factor: age distribution for U.S./Europe
  - superemitters
- off-road mobile/industrial
  - estimates by Kean et al. (2000)
  - need updates w/current measurements
- non-road transport
  - EPA documents (memos)
- "real world" factor?
  - not yet included



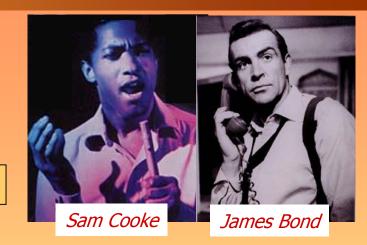
#### some "corrections" to earlier work

Bond (2004) vs Cooke (1999)

Differences are easily explained.

#### Coal, power generation (difference 1.5 Tg/yr)

We rely on measured BC fractions (<1%) instead of guesses (25%)



#### On-road diesel (difference 1 Tg/yr)

We use emission measurements and World Bank studies instead of assuming "developing countries have 5x higher emissions" (15 g/kg average PM emission factor)

#### Domestic diesel (difference 0.25 Tg/yr; large in Europe)

We do not apply emission factors for internal combustion engines to external-combustion boilers

# present dilemma

- "corrections" reduced emission estimates from 14 Tg/yr to 8 Tg/yr
- models typically need more BC to match observations... not less!

Need help from you! ("Real-world", source apportionment...)

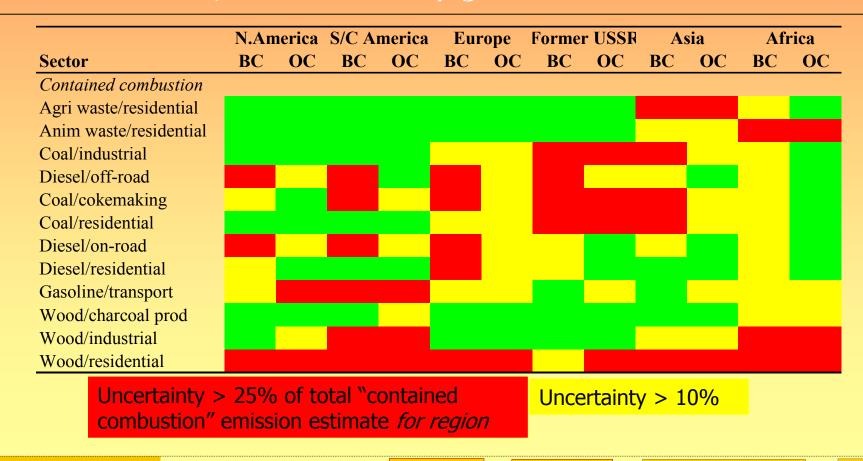
"What are the limits in our ability to measure freshly emitted and ambient BC?"

By most accounts, either thermally- or optically- measured BC is uncertain by a factor of 2.

This uncertainty affects our ability to corroborate emissionbased global transport models by using ambient measurements.

### uncertainties

- inventory contains full uncertainty propagation (activity estimates, emission factors, etc)
- of course, there are many guesses



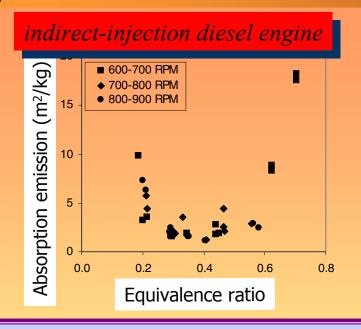
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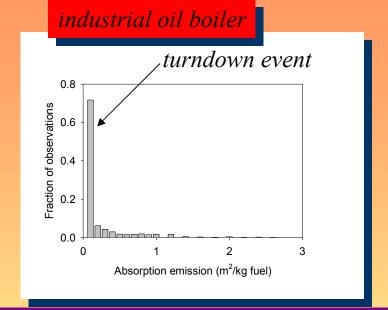
emission

concentration

impact

# single-source variability





"What are the limits in our ability to predict the mass of BC emissions from individual source types?"

Many of our sectoral emission estimates are uncertain by a factor of ~2. There is wide variability in both within-source and between-source variation.

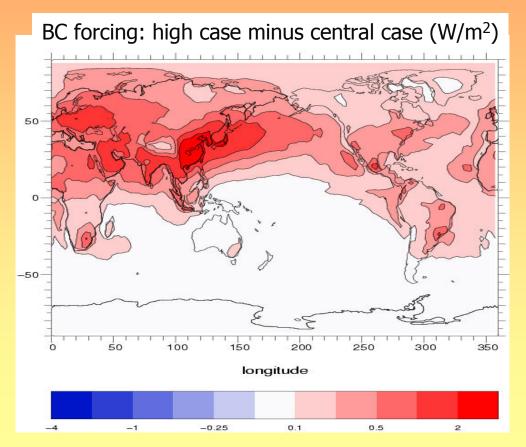
Note: diesel work is excellent forum for examining variability issues & developing approaches!



## emission uncertainty > forcing

graph & table represent spatially-dependent forcing uncertainty due to uncertainty in *fossil-fuel emissions* only

Model: NCAR-MATCH with 2001 NCEP-reanalysis met data



<b>TOA Forcing Summary</b>		
	BC	OC
Low	+0.26	-0.21
Central	+0.37	-0.22

+0.65

+0.40

High

Prev 84

-0.25

-0.23

### model intercomparison

why do <u>direct forcing</u> results differ?

#### black carbon

Chung, Haywood, Jacobson, Koch, Myhre, Penner, Wang

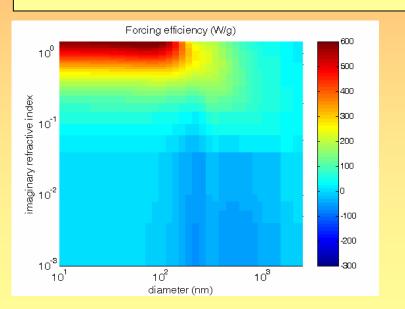
- lifetime
- optical assumptions
- vertical location

#### better resolved than OC:

- normalize by lifetime & optics
- TOA forcing estimates vary by 20%

#### organic carbon

- lifetime
- optical properties
   (absorp by OC) → factor-2
- water uptake ("f-RH")
- $\rightarrow$  factor-2



# accounting for OC properties

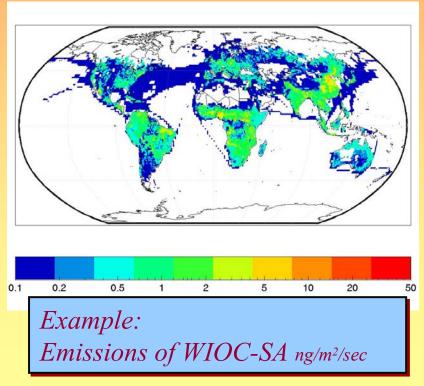
#### Haolin Sun, UIUC

- "OC" is dead; long live OC
- Climate-relevant, technology-specific OC divisions

#### **CROSS1-OC**

Climate
Relevant
Optical &
Structural
Subgroups
(ver 1) of OC

4 groups, tractable for climate models (we hope)

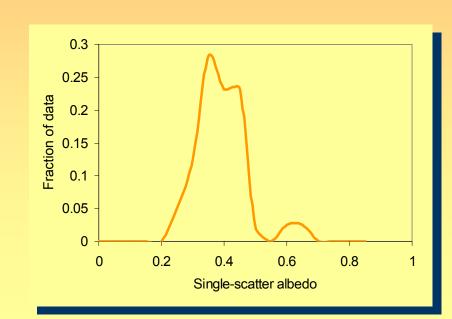


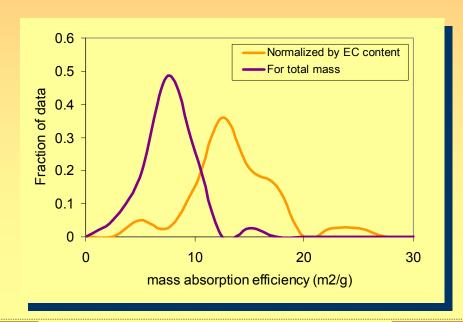
## climate-relevant source "profiles" (I)

- we can usually measure size
   need relationships for mobility of fractal particles
- ability to measure *relevant* composition needs work

light-absorbing component; other properties

#### Results of solid-fuel source tests

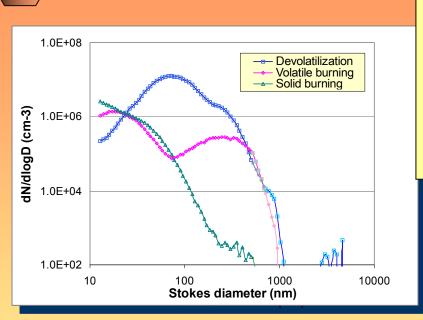




4. optics

action → emission → concentration → impact

# source "profiles" (II)



measured
size (SMPS/APS)
OC/EC
major ions
trace metals

measured absorption scattering

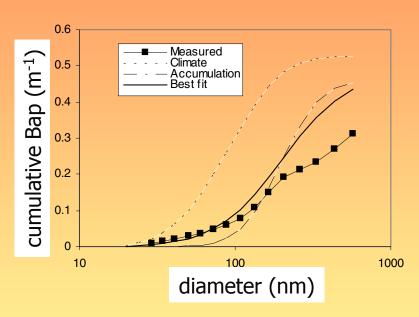
"closure" still difficult

"What are the limits in our ability to measure freshly emitted and ambient BC?"

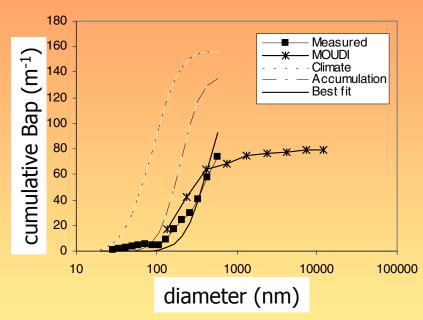
Because thermally-measured EC is uncertain (by x2?), we can't normalize absorption to an invariant quantity. Thus, it's difficult to corroborate models of optical transformation.

# different sources, different optics

#### 2 types of vehicles



A "nice" engine: small particles "look" like the BC that's in the models, larger particles appear less pure



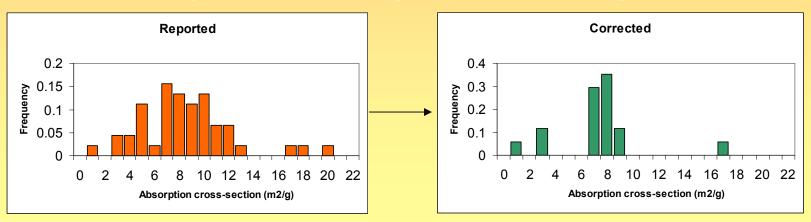
A "yucky" engine: much larger particles

# resolution & challenge

### For <u>fresh</u> light-absorbing carbon,

- we think we can explain
  - many variations in refractive index
  - variations in mass absorption cross-section
- + ...and yet,
  - "best guess" refractive index + theory doesn't match "best-guess" absorption cross-section

[Bond & Bergstrom, LAC investigative review, on verge of submission]



# rephrased question

"What are the limits in our ability to predict **the total forcing by aerosols** from individual source types?"

BC-OC and direct-indirect balances matter.

We need a clear identification of relevant factors. Optical properties and size are two of them. However, there are others.

Don't overestimate the present sophistication of global models. "In-situ" measurements are well ahead of the measurement-model link.



- Identify the invariant strongly light-absorbing quantity (and a method of measuring it)
- Statistical characterization of small-source populations (e.g. vehicles)
- Measured closure of optical and cloud-relevant properties from model variables, on fresh and transformed/transforming aerosol systems
- Identify key sensitive properties for determining direct & indirect forcing (from models)
  - iterate with inventories & ambient measurements
  - use urban/regional models to identify initial processing